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Traveling-wave deceleration of SrF molecules

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Abstract

We report on the production, deceleration and detection of a SrF molecular beam. The molecules are captured from a supersonic expansion and are decelerated in the $X^2\Sigma^+(v=0, N=1)$ state. We demonstrate the removal of up to 40% of the kinetic energy with a 2 meter long modular traveling-wave decelerator. Our results demonstrate a crucial step towards the preparation of ultracold gases of heavy diatomic molecules for precision spectroscopy.

1 Introduction

1.1 Motivation

Heavy diatomic molecules have a high potential as sensitive probes of parity violation [1, 2, 3]. The close proximity of opposite parity states offered by selected molecular systems causes a huge enhancement of weak interaction effects compared to atoms [4]. However, so far parity violation at atomic energy scales has only been studied in atoms, but not yet in molecules. One of the biggest hurdles is of experimental nature: methods have to be developed to control and prepare a sufficiently large number of molecules in a well-defined quantum state, as the starting point for any sensitive measurement.

Recently, successful experiments probing fundamental physics using the special properties of selected diatomic molecules have been performed using molecular beams, for example to put a limit on the possible size of the electric dipole moment of the electron [5, 6]. Molecular beams offer relatively high density, but limited interaction time. For a next generation of precision measurements using

heavy diatomic molecules we explore the possibilities to make use of a cold gas of trapped molecules. Trapped molecules potentially offer very long coherence times that can more than compensate for the lower number of molecules; such that weak interaction effects can be studied as perturbations on a coherently evolving superposition of molecular states.

A diverse range of techniques to decelerate, cool and trap molecules has been developed in the past decade. The motivation to create cold molecular gases ranges from the possibility to study the strong long-range dipole-dipole interactions [7], to molecular collision studies with an unprecedented level of control of the internal state of the molecules [8, 9], tests of fundamental physics such as the electron EDM as well as the study of possible variations of fundamental constants [10, 11]. More applications and an overview of the techniques to produce cold molecules have been addressed in recent review articles [12, 13].

The SrF molecule is a sensitive candidate to study weak interaction effects, specifically nuclear-spin dependent effects and the anapole moment of the nucleus. Coincidentally, the SrF molecule is prototypical of a group of alkaline-earth monohalide molecules whose properties allow for a certain amount of laser cooling due to favorable Franck-Condon factors. Molecular laser cooling was first demonstrated to work for SrF [14], and other suitable molecules include YO [15], CaF [16] and probably YbF [17] and RaF [18]. However, since the transitions used are not completely closed due to residual leaks to excited vibrational levels, it is challenging to provide a sufficiently strong force by scattering photons to completely cool and trap the molecules (even from a pre-cooled molecular beam).

Due to its sizeable dipole moment of 3.5 Debye, the lowest rotational levels of the SrF molecule in its $^2\Sigma^+(v=0)$ absolute ground state have a reasonable Stark shift in external electric fields, and are therefore amenable to Stark deceleration. Our approach to create ultracold samples of SrF molecules is to combine a traveling-wave Stark decelerator with molecular laser cooling. The decelerator can capture an internally cold sample from a supersonic expansion and bring this sample to rest in the laboratory frame. By scattering ~ 1000 photons the sample can be brought to the Doppler temperature limit. As the molecules remain trapped during the entire deceleration process, losses of slow molecules due to transverse beam spreading are absent.

In this article we describe the creation of a supersonic beam of SrF molecules, the traveling-wave guiding and deceleration of these molecules and the detection of the decelerated molecules using laser induced fluorescence. We demonstrate all key components needed to fully stop and statically trap a pulsed beam of SrF molecules. For the measurements presented here a modular decelerator of 2 meter length has been employed; for complete stopping this decelerator has to be extended to a length of 5 meter. The good agreement of our measurements with the numerical simulations that we have performed demonstrates that traveling-wave decelerators of this length are indeed completely stable: molecules that fall within its phase-space acceptance are guided or decelerated without losses.

1.2 Stark deceleration of heavy diatomics

Stark deceleration uses the fact that the energy levels of molecules with an electric dipole moment shift in an external electric field. When such a molecule travels through an inhomogeneous electric field it loses kinetic energy while it gains potential energy from climbing up the electric field hill. If this process of climbing the potential hill is repeated many times, the kinetic energy loss can be considerable. In practice this is done for light molecules in a traditional Stark decelerator by repeatedly switching the polarity of an array of high-voltage electrodes through which the molecular beam traverses. In comparison to lighter molecules with a linear Stark shift, heavy diatomics such as SrF are more difficult to decelerate. First, for a fixed initial velocity and deceleration strength, the large mass requires a rather long decelerator. This puts high demands on the stability of the deceleration process [19]. Second, related to the small spacing of rotational levels, at modest electric fields (20-30 kV/cm) the slope of the Stark curve of the lowest rotational levels turns negative; at higher fields these states are all so-called high-field seekers, and are attracted to the high field at the electrodes. The deceleration of molecules in high-field seeking states has proven to be very challenging [20]. At the turning point the maximum Stark shift for the lowest rotational states is only a fraction of a wavenumber. This is illustrated in Figure 1 for the lowest rotational states of SrF in its $X^2\Sigma^+(v=0)$ electronic and vibrational ground state. The unfavorable Stark shift and the high mass result in highly inefficient deceleration of SrF with a traditional Stark decelerator. Therefore we have built a traveling-wave decelerator [21] consisting of a sequence of ring electrodes which are used to propagate 3D electric traps that can hold bunches of molecules, thereby mitigating most of the losses that traditional Stark decelerators have. Motivated by similar arguments, an experiment has been performed on the deceleration of YbF molecules in a 0.5 meter traveling-wave decelerator [22]. Recently the static trapping of ammonia molecules in a traveling-wave decelerator has been demonstrated [23, 24].

The working principle of the traveling-wave decelerator has been discussed in literature [21, 25, 26]. Here we briefly summarize the essential features. A sequence of ring-shaped electrodes is supplied with a sinusoidal voltage that varies in space and time such that there is a phase shift of 45 degrees between consecutive electrodes. Electric field minima and maxima are created along the axis of the ring structure as shown in Figure 2. Inside the periodic structure, the electric field minima will appear at regular intervals. These minima can be moved along the decelerator axis by applying these oscillating voltages to the rings in such a way that a traveling wave is created. Molecules in low-field-seeking states will experience these minima as three-dimensional traps. The speed of the traps can be changed by varying the frequency of the oscillating voltage.

Here we report first time deceleration of SrF molecules. We show that the modular traveling-wave decelerator of 2 meter length (4 modules of 0.5 meters each) is stable in operation and construction. This is of crucial importance for the extension of the device to ~ 5 meter length which is required to completely

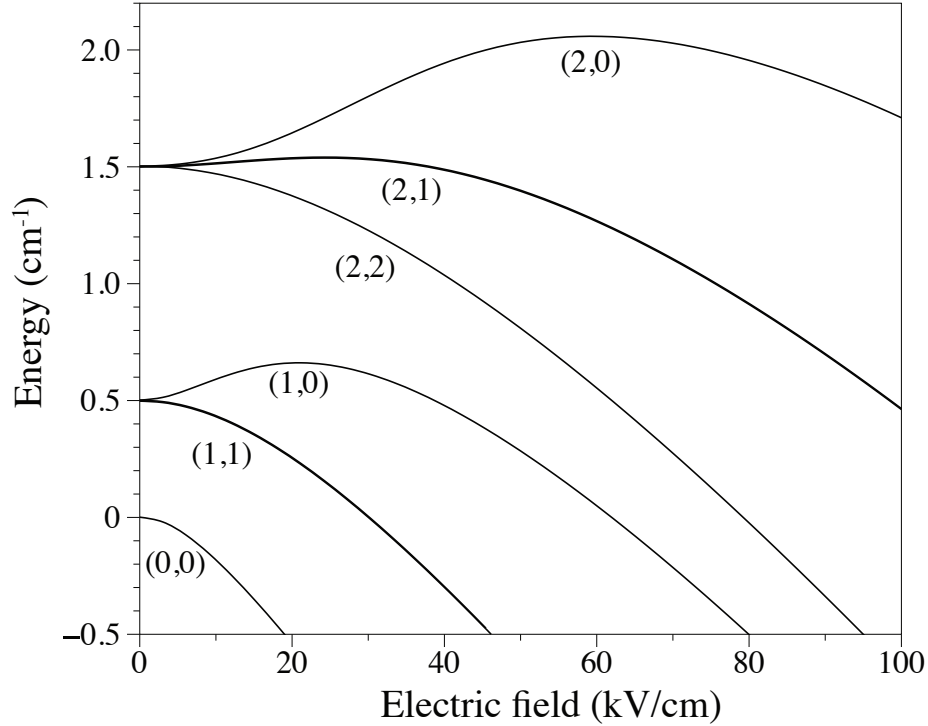


Figure 1: The Stark shift of the three lowest rotational levels of the ^{88}SrF molecule in its $X^2\Sigma^+$ groundstate. The levels are labeled (N, M_N) where N is the rotational number and M_N the projection of N on the electric field axis. Hyperfine structure is not visible on this scale.

stop SrF molecules.

2 Experimental setup

Figure 2 shows a schematic view of our deceleration experiment consisting of a supersonic expansion source, the decelerator and a laser-induced fluorescence detection zone. A 100 mJ pulse from a Nd:YAG laser at its principal wavelength is used to ablate SrF radicals from a home-pressed pill consisting of 90% SrF_2 and 10% B into a supersonic expansion of Xe gas from a pulsed General Valve. The addition of boron is found to make the pill much more stable [27]. The valve is operated at 10 Hz and it is cooled with cold nitrogen gas to -30°C , resulting in an average molecular beam speed of 300 m/s. From a rotational spectrum we find a rotational temperature of ~ 10 K. Upon optimisation of the source we expect to be able to reach a comparable output as a supersonic source for YbF molecules [28]. The molecular beam enters the decelerator located 125 mm

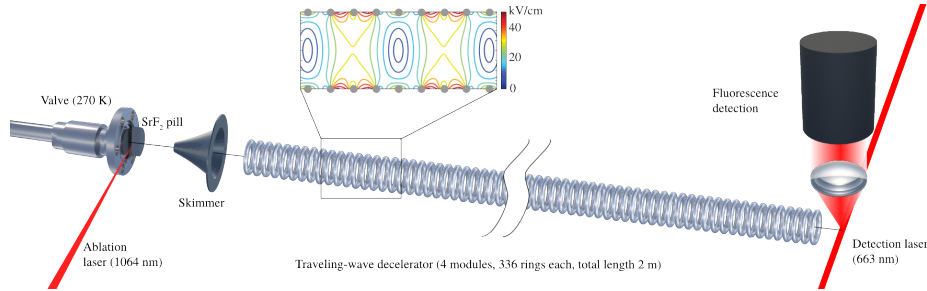


Figure 2: A schematic view of the experiment. SrF molecules are created by laser ablation from a pill. A pulsed supersonic Xe expansion cools the SrF molecules and takes them to the ring structure where the molecules are guided or decelerated. Further downstream the SrF is detected by laser induced fluorescence.

downstream of the valve through a skimmer with a diameter of 2 mm, placed 60 mm from the ablation spot.

The dimensions of the ring-shaped electrodes and the method in which these are positioned relative to each other is based on the design of Meek *et al.* [25]. Since we require a rather long decelerator however, we combine four modules of 50 cm in order to form a horizontally oriented decelerator with a length of 2.016 m. Each module contains 336 ring-shaped electrodes made of tantalum wire of 0.6 mm diameter. The electrodes are mounted on eight 8 mm diameter cylindrical stainless steel rods which are placed in an octagonal pattern on the outside of a circle with circumference of 26 mm. The gap between two consecutive rings is 0.9 mm, resulting in a periodicity of 12 mm.

2.1 High-voltage electronics

Arbitrary waveforms are generated using DACQ8150 Acquiretek PCI cards and amplified by eight custom high-voltage amplifiers from Trek Inc. These amplifiers have an output voltage of maximally ± 5 kV at frequencies between 30 kHz and DC and can handle the full capacitance of a 5 meter long decelerator. This makes it possible to decelerate molecules to a complete stop and then keep the molecules in a static trap. Furthermore, the arbitrary waveforms can be used to tailor the shape of the potential at will or to manipulate the trap as we like [24].

2.2 Detection

After deceleration and 116.5 mm of free flight, the molecules are state-selectively detected using a resonant laser-induced fluorescence scheme. Light at 663.3 nm to drive the $A^2\Pi_{1/2}(v=0) \leftarrow X^2\Sigma^+(v=0) P(1/2), Q(1/2)$ transitions, which probes the population in the lowest low-field seeking $N = 1$ rotational level, is generated using an external-cavity diode laser. It is this state which is the

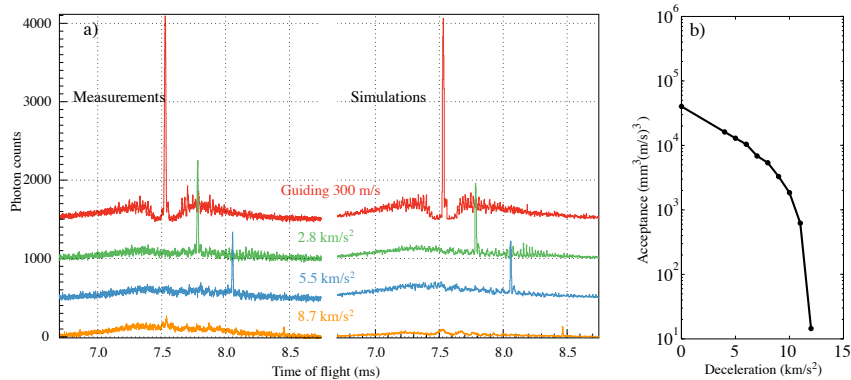


Figure 3: a) Measured (left) and simulated (right) time-of-flight profiles for SrF (1,0) molecules after 2 m of deceleration. The different measurements have been given a vertical offset for clarity. b) The 3D acceptance of the traveling-wave decelerator as a function of deceleration strength, as found from numerical trajectory calculations [26].

starting point for a parity violation measurement, and the state for which laser cooling has been demonstrated to work. The $N = 1$ state is also the best choice for deceleration given the electric field strength available to us. Approximately 10 mW of laser light is used to lock the laser to absorption line R(115) (6-6) a9 in molecular iodine enclosed in a quartz cell heated to 300 °C. We identify this line using frequency-modulated doppler-free saturated-absorption spectroscopy. The light used for SrF detection is sent through an EOM driven at 41 MHz for the creation of sidebands that overlap with the resolved hyperfine levels in the SrF $X^2\Sigma^+(v = 0, N = 1)$ state [14]. It reaches the detection chamber through a single mode optical fiber where it is directed through the detection region using two mirrors. The beam is retro-reflected such that it crosses the molecular beam at right angles twice. Typically, the laser power is 2 mW and the beam diameter is 3 mm. A system of lenses focusses the fluorescent light through an interference filter onto the cathode of a photomultiplier tube (PMT). We have an overall detection efficiency of 2%. The arrival time of the PMT pulses is recorded by a multi-channel analyzer with 1 μ s bins. The time-of-flight spectra are started with the ablation pulse.

3 Results

Here we present the results obtained from guiding and decelerating SrF(1,0) molecules with the traveling wave decelerator.

The left side of Figure 3a shows measured time-of-flight (TOF) spectra of SrF(1,0) molecules. For each spectrum a small constant background has been subtracted. The spectra have been given a vertical offset for clarity. The mean

speed of the beam was 300 m/s and the starting speed for the decelerator waveforms was set accordingly. The arrival time of the molecules is plotted for guiding at 300 m/s and a range of deceleration strengths, each of which was constant throughout the full 2 m length of the decelerator. The delayed arrival of the main package of molecules with increasing deceleration strength can clearly be seen. At the highest deceleration of 8.7 km/s^2 the package of decelerated molecules is arriving around 8.45 ms, corresponding to molecules that were decelerated from 300 m/s to 234 m/s. This corresponds to a removal of 40% of the kinetic energy.

The TOF profiles show very distinct features which are best seen in the guiding measurement (Figure 3a upper trace). At 7.5 ms there is a sharp peak that corresponds to molecules that are trapped in a moving electric trap. The small maxima next to the main peak are due to molecules that were trapped in potential wells adjacent to the central well. Because of the small size of the SrF cloud at the entrance of the decelerator almost all SrF molecules are loaded into a single potential bucket. The molecules that are collected in these wells leave behind a big dimple in the final TOF spectrum which is broadened because of flight time. The molecules that fall outside the longitudinal phase-space acceptance but remain inside the transverse phase-space acceptance of the decelerator are not captured in the potential well, but are still guided through and detected. These molecules end up in the broad wings that are observable in the guiding signal around 7.3 and 7.8 ms. The modulation that is observable on these broad wings is caused by bunching in phase space on the moving potential hills in between the electric field minima.

We have calculated [26] the phase space acceptance of the decelerator for different deceleration strengths as shown in Figure 3b. As an addition to this we have now simulated the TOF spectra as well, including the effects of the shape of the initial distribution. By comparing the simulations to the measurements, we derive the properties of the molecular beam after the supersonic expansion. We find a longitudinal velocity distribution with a mean speed of 300 m/s and a standard deviation of 20 m/s. Furthermore the starting size of the molecular packet is of order 1 mm, which matches to the spot size of the ablation laser on the pill. As displayed on the right side of Figure 3a all the features of the measurements are reproduced in the simulations. The smaller peaks for higher deceleration strengths can be understood from the decreasing phase space acceptance and are not due to losses in the decelerator. The simulations have been scaled on the guiding signal to match the intensity of the non-decelerated part of the detected molecules. The height of the decelerated peak in the simulation for the strongest deceleration is higher than measured, which we attribute to suboptimal experimental conditions. During the experiments the laser ablation power gradually reduced and the poor regulation of the valve temperature caused the supersonic expansion to change slightly. The experiment with the strongest deceleration is the most sensitive to these effects, because of the increased sensitivity to the timing of the molecular pulse relative to the start of the deceleration, since the phase-space acceptance is rather small. As a consequence of these measurements we are implementing an improvement

of the temperature regulation of the cooled supersonic expansion.

Each spectrum shown is the sum of 10240 ablation shots, except for the strongest deceleration strength which is the average of twice as many shots because of frequent unlocking of the spectroscopy laser. In a single shot only a few photons are detected, indicative of a rather low beam density. Being cautious not to damage to the ablation pill, we have used the lowest possible ablation power that enabled us to still measure with a good signal-to-background ratio. Obtaining the time-of-flight traces in Figure 3 took about 20 minutes per trace. In later experiments we have seen that the ablation power could be increased significantly without damaging the pill, giving a correspondingly higher yield of SrF molecules. We are currently optimizing the ablation geometry to provide better rotational cooling of the molecular beam, thereby increasing the fraction of molecules in the (1,0) state.

4 Conclusions

Here we report the first Stark deceleration of SrF molecules. These measurements demonstrate the successful combined operation of the supersonic expansion source of SrF molecules, the 2 m long modulator decelerator and the laser-induced fluorescence detection. Extension of the decelerator to 5 m is underway, and it is expected to deliver completely stopped bunches of SrF molecules. Such a decelerator is a general device, extending the range of molecules that can be decelerated and trapped. These include relatively heavy molecules with an unfavorable Stark shift such as SrF, YbF and PbO, larger (bio)molecules such as benzonitrile [29], but also lighter molecules that have so far eluded Stark deceleration such as water. We expect these cold samples of molecules, especially in combination with further cooling methods, to be the starting point for many exciting future experiments.

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